The NIST high-flux backscattering spectrometer

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Abstract. The neutron-backscattering technique extends the dynamic range of neutron time-of-flight and conventional triple-axis spectrometers down to μeV energies and is ideal for the study of slow motions in complex liquids, jump diffusion, and quantum rotational tunneling. We report on the performance of the new high-flux backscattering spectrometer (HFBS) at the NIST Center for Neutron Research. Compared to other backscattering spectrometers, the HFBS achieves a higher neutron intensity through the use of a device called a phase space transformation chopper and a large analyzer array. A custom-designed Doppler drive for the monochromator provides a triangular velocity profile capable of high-frequency operation, thus extending the dynamic range of the spectrometer by more than a factor of two over similar instruments.

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Cold neutron backscattering spectroscopy is an important tool for studying slow $(0.1 < f\,(\text{GHz}) < 10)$ molecular and atomic motions in solids and liquids. The typical energy resolution of a backscattering spectrometer can reach below 1 μeV, corresponding to times of order 1 ns. Thus it is possible to probe dynamics with backscattering spectroscopy on much longer time scales than is possible with the cold neutron time-of-flight technique for instance. These time scales are important for studying polymer and protein dynamics, dynamics of viscous liquids, tunneling motions, and dynamics of guest molecules in various confining geometries.

A conventional neutron backscattering spectrometer achieves an excellent energy resolution by exploiting the fact that the wavelength spread of neutrons scattered from a crystal is minimized when the scattering angle is 180°. Backscattering is used both to monochromate the incident beam and to energy analyze the beam scattered from the sample. An unfortunate consequence of this high resolution technique is

low flux. In order to make the backscattering technique feasible, the analyzer system usually subtends a large solid angle, thus degrading the Q-resolution and creating a rather large beam divergence mismatch between the incident and scattered neutron beams. The HFBS incorporates a number of neutron optical devices in order to reduce this mismatch and enhance the flux at the desired backscattering energy. In this paper we describe the design and performance of the HFBS at the NIST Center for Neutron Research.

1 Instrument design and performance

The primary goals in the design of the HFBS were to obtain a high flux on the sample, a large scattered flux in the detectors, a large dynamic range, and sub-μeV energy resolution. To this end a number of critical features were incorporated into the design of the instrument. Some details of the instrument design have been presented elsewhere [1], and a more detailed discussion is in preparation [2], but we provide here a very brief summary of the novel design components.

The instrument, as shown in Fig. 1, can be functionally decomposed into a primary and secondary spectrometer. The primary spectrometer consists of the neutron guide system, a velocity selector, a beryllium-bismuth filter, a converging guide, a phase-space transformation chopper (PST), and the Doppler monochromator. The secondary spectrometer is composed of a large spherically curved analyzer system (composed of large diameter Si(111) hexagons) subtending 20% of a solid sphere and 16 ^{3}He detectors spanning 14.46° to 121.25° in scattering angle.

The main practical limitation of the neutron backscattering technique is the low flux due to the selection of such a narrow spread in both incident and final neutron energy. The HFBS incorporates a novel device known as a phase space transformation chopper that effectively enhances the neutron flux at the backscattering energy (2.08 meV) of the monochromator (at rest) and the analyzer system [3]. This comes at the expense of a larger horizontal divergence, but this divergence more closely matches the divergence of the analyzer-detector system than would operation without

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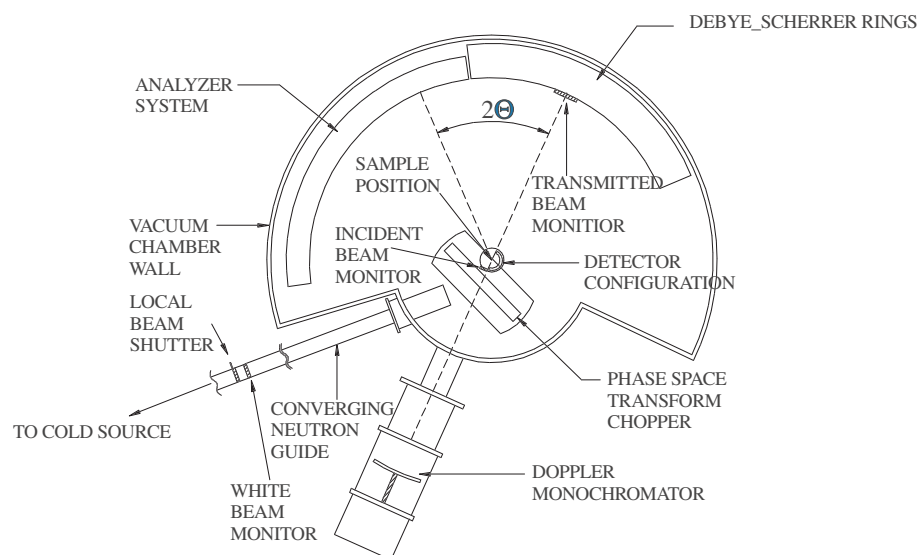(Fax: +1-301/9219847, E-mail: robert.dimeo@nist.gov)

the PST. The PST provides a fourfold gain in intensity at 2.08 meV, resulting in a flux at sample position of $1.4 \times 10^5 \, \mathrm{n \, cm^{-2} \, s^{-1}}$. To our knowledge, the HFBS is the only neutron spectrometer in the world currently using a PST but the design for new backscattering instruments currently under development at other facilities includes a PST [4].

The Doppler monochromator system employs a motor-driven cam that has been machined to yield a velocity profile that is approximately triangular. This provides almost equal weight to all energy transfers in the dynamic range. Although the Doppler monochromator system was designed to operate at any frequency up to 25 Hz (corresponding to an energy transfer of $\pm 50 \, \mu\mathrm{eV}$), there are a number of frequency intervals that are forbidden due to mechanical resonances that produce a significant distortion of the lineshape. Thus, the HFBS is routinely operated with the following dynamic ranges and energy resolutions (full-width at half maximum): $\pm 11 \, \mu\mathrm{eV}$, $\delta = 0.79 \, \mu\mathrm{eV}$; $\pm 20 \, \mu\mathrm{eV}$, $\delta E = 0.87 \, \mu\mathrm{eV}$; $\pm 36 \, \mu\mathrm{eV}$, $\delta E = 1.01 \, \mu\mathrm{eV}$.

2 Examples

The large dynamic range, excellent energy resolution, and high flux of the HFBS allow the study of a number of interesting systems. Examples include tunneling in molecular solids and systems undergoing structural relaxation. We present three examples of recent measurements performed with the HFBS.

Toluene ($C_6H_5CH_3$) has two known crystalline phases: the α phase and the β phase with respective melting points of $T_{\mathrm{melt}}(\alpha) = 178 \, \mathrm{K}$ and $T_{\mathrm{melt}}(\beta) = 154 \, \mathrm{K}$. In the α-crystallized molecular solid there are two inequivalent methyl groups (CH_3) that give rise to two rotational tunneling lines on each side of the elastic line, all of which are observable with neutron backscattering [5]. In Fig. 2 we show a spectrum taken at 15 K and summed over a wavevector transfer range of $0.56 \, \text{Å}^{-1}$ to $1.6 \, \text{Å}^{-1}$ [6]. With a Doppler monochromator frequency of 16.7 Hz the dynamic range of the HFBS is $\pm 36 \, \mu\mathrm{eV}$ and the energy resolution is $1 \, \mu\mathrm{eV}$. The spectrum in Fig. 2 clearly shows the large dynamic range and excellent

energy resolution necessary to resolve both pairs of tunneling peaks. To our knowledge, there is no other neutron spectrometer capable of resolving all four tunneling peaks in a single measurement.

A topic of current interest is the structural relaxation in polymer systems. A recent measurement of the glass-former polystyrene (PS) in a solution of d-toluene (15% concentration PS by molecular weight) revealed clear non-exponential structural relaxation. A spectrum collected at $0.99 \, \text{Å}^{-1}$ and $T = 220 \, \mathrm{K}$ is displayed in Fig. 3 [7]. The data were modeled with a Kohlrausch spectral function (the Fourier transform of a stretched exponential function),

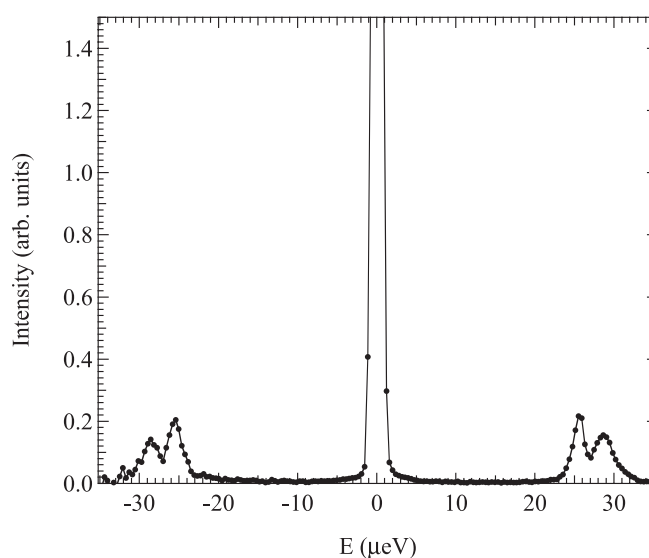$$S(Q, \omega) = \frac{1}{\pi} \int_0^\infty \cos(\omega t) \exp\left(-\left(\frac{t}{\tau} \right)^\beta \right) \mathrm{d}t \,. \tag{1}$$

Fig. 2. Tunneling spectrum of toluene at $T = 15 \, \mathrm{K}$ and summed over a wavevector transfer range of $0.56 \, \text{Å}^{-1}$ to $1.6 \, \text{Å}^{-1}$. Instrumental resolution is approximately gaussian with a full-width at half maximum of $1.0 \, \mu\mathrm{eV}$. Solid line is a guide to the eye
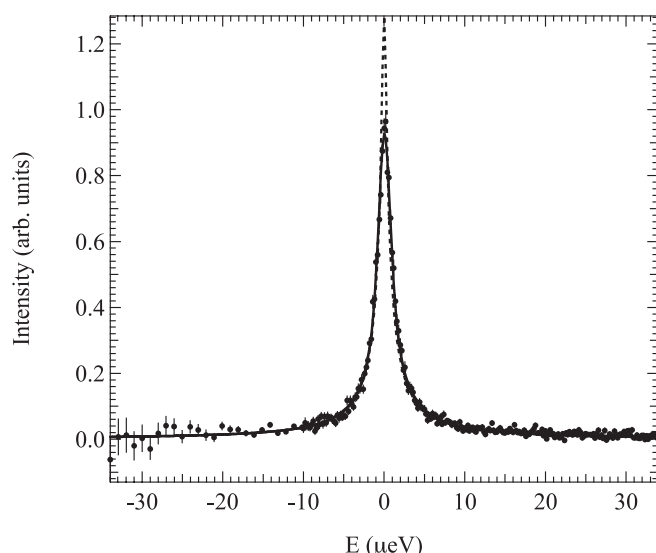
Fig. 3. Structural relaxation of polystyrene in d-toluene at $T = 220$ K. *Solid line* is a fit to the Kohlrausch spectral function convoluted with the instrumental resolution function. *Dashed line* is the unbroadened Kohlrausch spectral function

Fig. 4. Effective mean square displacements for a 75 Å PC film (*solid circles*) and bulk PC (*solid line*)

Because the data have particularly large wings and are quite narrow near $\omega = 0$, this example illustrates the need for a large dynamic range and excellent energy resolution in order to extract reliable estimates of the stretch factor, β, and the relaxation time, τ.

The last example we present is from a recent measurement of the elastic scattering intensity as a function of temperature for bulk polycarbonate (PC) and that from a 75 Å PC film deposited onto the surface of silicon wafers via spin-coating [8]. These measurements are performed with the Doppler monochromator at rest and are referred to as fixed window scans. Assuming that the scattering arises from a solid, the mean-squared displacement of the scatterers can be extracted from the Q and T-dependence of the elastic scattering intensity. The result of such an analysis is shown in Fig. 4. The effect of the restricted geometry of the confined PC is clearly shown by the smaller values of the mean squared displacements. Such measurements as this one can provide valuable information on the effects of restricted geometry on polymer dynamics. However these measurements can be very difficult because there is so little sample. The sample
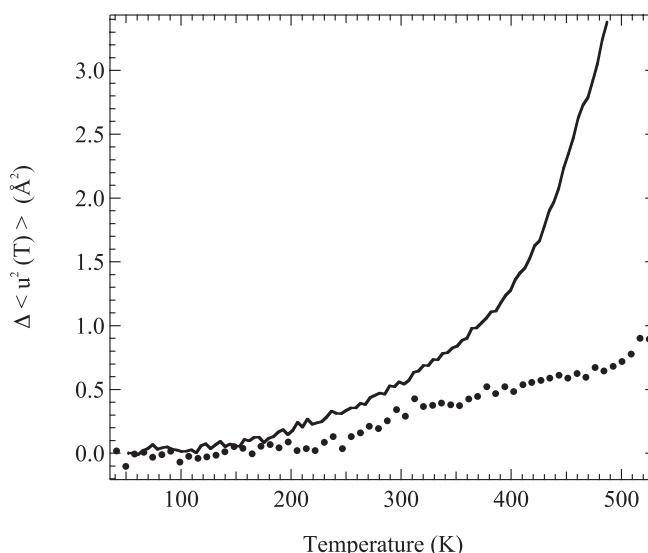
consisting of the 75 Å PC film, deposited onto enough wafers to fill the beam, contained only 0.56 mg of PC and the count time was 10 min K^{-1} (data has been rebinned in Fig. 4). Thus the high flux is absolutely essential for performing measurements on such dilute systems.

The HFBS at the NIST Center for Neutron Research is a high-performance neutron backscattering spectrometer with high intensity, large dynamic range, and excellent energy resolution.

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